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FINAL REPORT

for

Contract No. NAS8-31538

"Preliminary Aerosol Generator Design Studies"

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This report was prepared by the University of Missouri-Rolla, Cloud Physics Research Center, under Contract No. NAS8-31538, "Preliminary Aerosol Generator Design Studies", for the George C. Marshall Space Flight Center of the National Aeronautics and Space Administration.

I. INTRODUCTION

The scope of the work to be performed under this contract was "to accomplish the preliminary design and prototype construction of an aerosol generating system for use in the Science Simulator of the Cloud Physics Laboratory Project and also as part of the Cloud Physics Laboratory payload to be flown on the Shuttle/Spacelab. Both atomization and vaporization techniques should be investigated".* The original proposal submitted to NASA placed primary emphasis on an atomization type generator to produce moderately large insoluble particles with secondary emphasis on vaporization techniques for small soluble aerosols. Even before work started on this contract it became evident that the most pressing need was for a highly dispersed aerosol; that is, an aerosol of very small particle size. More specifically the requirements were for a NaCl aerosol with a size distribution such that the ratio of the concentration of particles with diameters between 0.02 and 0.1 μm was at least 500 times the concentration of particles with diameters greater than 0.1 μm . It also soon became evident that such an aerosol could not readily be produced using atomization techniques. Thus the work performed under this contract has been directed toward the design and construction of a prototype vaporization generator for highly dispersed sodium chloride aerosols.

In summary, we have built four generators which will produce the required aerosols. Unfortunately, we have not yet

*Exhibit "A"; Scope of Work, Contract NAS8-31538.

found it possible to predict in advance the distribution which will be obtained. Further, the distribution often varies slowly with time. We believe that with further development it might well be possible to improve the stability as well as having a higher probability of being able to predict the size distribution which will be obtained. Finally, we present the results of two coagulation experiments. It may well be that the only way to obtain an aerosol which, if not stable, for the required length of time will at least change in a reproducible manner, is by filling a storage container from which the aerosol is removed when needed.

II. GENERATOR DESIGN PHILOSOPHY

There are two principal methods by which aerosols are generated: aerosolization and condensation.

A. Aerosolization.

When a bulk, condensed phase is broken up into smaller particles, relatively large amounts of energy must be added to the system because of the large increase in surface area of the dispersed particles as compared to the original bulk material. Because of this, it is impractical to attempt to produce particles by aerosolization, particularly with solids, with sizes approaching a few tenths of a micrometer. Theoretically it would be possible, particularly with liquids, to produce larger particles of pure materials by aerosolization and then reduce their size by controlled evaporation. Practically, however, controlling the extent of evaporation to produce a given size aerosol would be difficult. Also, nonvolatile impurities in the starting material could produce nuclei with a high percentage of impurity concentration (see below).

A more common procedure is to generate an aerosol from a solution of the desired nonvolatile aerosol substance in a volatile solvent. Evaporation of all the volatile solvent will then leave the nonvolatile solute as a residue. Again there is a practical difficulty. Assuming 1 μm diameter liquid drops are produced, if the concentration of nonvolatile impurities in the solvent is 5×10^{-2} v/o (a not unreasonable figure) a residue of 0.08 μm diameter is left when all the

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solvent evaporates even with no added solute. Unfortunately, these impurity residues are in the range of sizes of the aerosol which need to be produced. The extent of this difficulty may be appreciated by noting that to reduce the size of the impurity particle by a factor of two would require either a starting liquid aerosol of $0.5 \mu\text{m}$ or an impurity concentration of $6.5 \times 10^{-3} \text{ v/o}$. Although not impossible, either of these requirements would be difficult to achieve and the size of the residue aerosol would still be $0.04 \mu\text{m}$.

B. Condensation

There are two methods by which aerosols may be produced by condensation: reaction or decomposition of volatile materials with subsequent condensation of the less volatile reaction products or vaporization of less volatile materials with subsequent condensation at a lower temperature.

Reaction or decomposition has been used to produce very small, highly dispersed, aerosols. Unfortunately the composition of the aerosols is limited to systems for which appropriate reactions or decompositions are available. For this reason, it has been used primarily with volatile metal compounds to produce metal or metal oxide particles and with organic compounds for the production of carbon black. While a reaction between sodium vapor and chlorine gas would probably produce sodium chloride aerosols, this appeared to be an impractical procedure at best.

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Vaporization with subsequent condensation can be used with any material which does not decompose under heat and has a sufficiently high vapor pressure at some temperature which can be readily attained. This is probably the most popular method for the generation of very small aerosol particles and has been used with metals, liquids and ionic solids.

While the specifics of this technique vary depending, at least in part, on the aerosol being generated, the general principles remains the same. A gas phase, containing the vapors of the aerosol material, is cooled to such a temperature that supersaturation results and the aerosol substance condenses. The gas phase can be cooled by adiabatic expansion, heat exchange with a colder body, mixing with a colder gas or a combination of these. Because for our purposes NaCl aerosols are desired, we consider below the generation of these aerosols.

In general, to prepare a NaCl aerosol by the evaporation-condensation method, solid NaCl is heated in a stream of dry gas. This gas is subsequently cooled in a condenser section either by heat conduction to the cooler walls of the condenser or by mixing with a cooler gas. A second furnace, hotter than the first, is often added downstream of the condenser to vaporize the aerosol which was formed in the condenser. The aerosol is then reformed in a second condenser section. The purpose of the second heater is twofold: the final aerosol often appears to be less polydisperse than without it and it allows some control over the rate at which the vapor laden air is cooled. To promote the condensation of the NaCl vapor, an

external source of condensation nuclei (CN) is often incorporated.

The mean size, dispersion and concentration of the final aerosol will depend on the concentration of NaCl vapor, how the gas is cooled and how nucleation proceeds. Theoretically, if a given volume of gas with a homogeneous distribution of NaCl vapor and identical condensation nuclei could be uniformly cooled at a rate which would not allow homogeneous nucleation or condensation of the salt on the walls of the container, a monodisperse aerosol should be produced with a concentration equal to that of the condensation nuclei and size determined by the concentration of NaCl vapor. In practice this ideal system is not realized due to the difficulty of cooling the air in such a manner that temperature and vapor concentration inhomogeneities do not occur.

In designing our generator for highly dispersed NaCl aerosols we have followed the general outline described above. In all, four generators have been built. In each case two heated sections were employed. Approximately 1 lpm of dry air was passed through the furnaces and cooled by mixing with 20 lpm of dry air immediately upon exiting the second heated section.

III. AEROSOL GENERATION SYSTEM

Figure 1 is a schematic diagram of the final aerosol generation system. For ease in describing the development and final design, the system has been divided into four subsystems: generator, temperature measurement and control, aerosol analyzer, and diluter.

A. Generator

This subsystem consists of the furnaces, salt source, and the input and dilution air. Four different generators have been constructed which will be described in chronological order.

The first generator, which is still used periodically to produce aerosols for the Graduate Center for Cloud Physics Research (GCCPR) Cloud Simulation Chamber, consists of two, 32 cm long x 3.2 cm I.D., Hoskins tube furnaces. These furnaces are placed, in-line, 8 cm apart which provides a condenser section between the two heated sections. A 3.2 cm O.D. x 2.5 cm I.D. mullite tube is passed through these furnaces. The salt source, a porcelain boat containing loose, crystalline NaCl, is placed in the middle of the first furnace. Air, at one lpm, is passed through the mullite tube. This air, the input air, is controlled with a rotameter flow meter. The exhaust end of the mullite tube is reduced to a 1 cm diameter tapered neck which is pressed into a 1 cm I.D. copper tee. The NaCl laden input air is mixed in this tee with 20 lpm of dilution air, also controlled with a rotameter flowmeter. Both input and dilution air are supplied from the same air preparation

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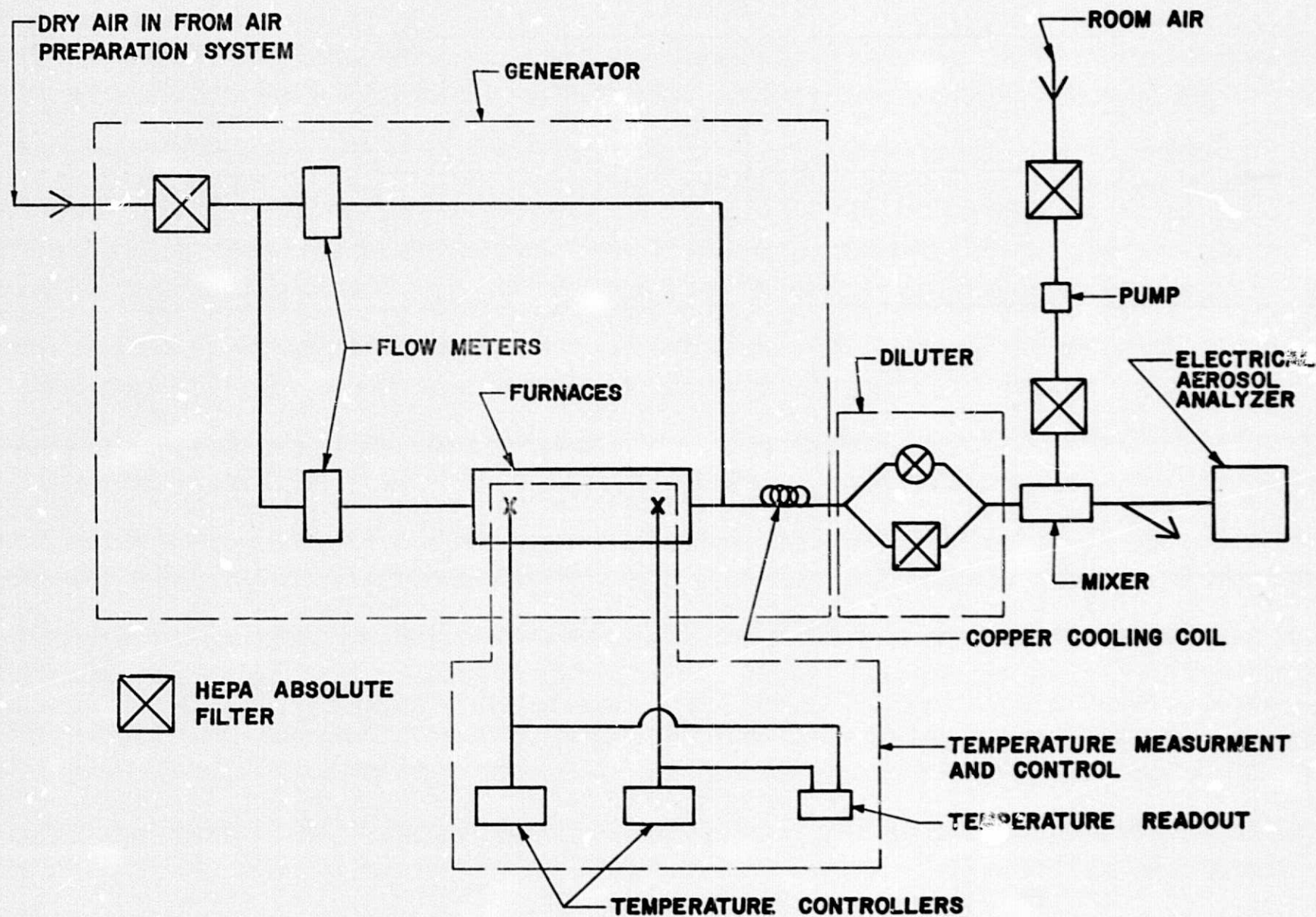


Figure 1.

system which supplies air for the GCCPR Cloud Simulation Chamber. This air is usually dried to a dewpoint of -20°C or less. The input and dilution air arrangement has remained essentially unchanged throughout this program.

This generator has proven quite successful in that it will produce the proper aerosol and remain stable for relatively long periods of time as shown in figure 2. In this case, the furnaces had been on for 45 hours before the data for curve number 1 were obtained. The approximate furnace temperatures when these data were taken were 750° in the first furnace with the salt source and 870° in the second furnace. This generator was originally constructed to provide preliminary information on the proper operating parameters. Unfortunately, the weight, size, power consumption and source configuration make it impractical for use with the Shuttle/Spacelab although it could be used with the Science Simulator.

The other three generators were all fabricated in the following general manner. Chromel A heater wire, 20 gauge, was wrapped on a 2.5 cm I.D. alundum heater core. A few layers of asbestor paper were wrapped around the wound core to provide electrical insulation between the heater winding and sensing thermocouples. These cores were suspended in metal cans by transite end plates. (Figure 3 shows the final prototype.) Fibrefrax insulation was packed tightly into the region between the core and the outside shell. Metal end caps held the

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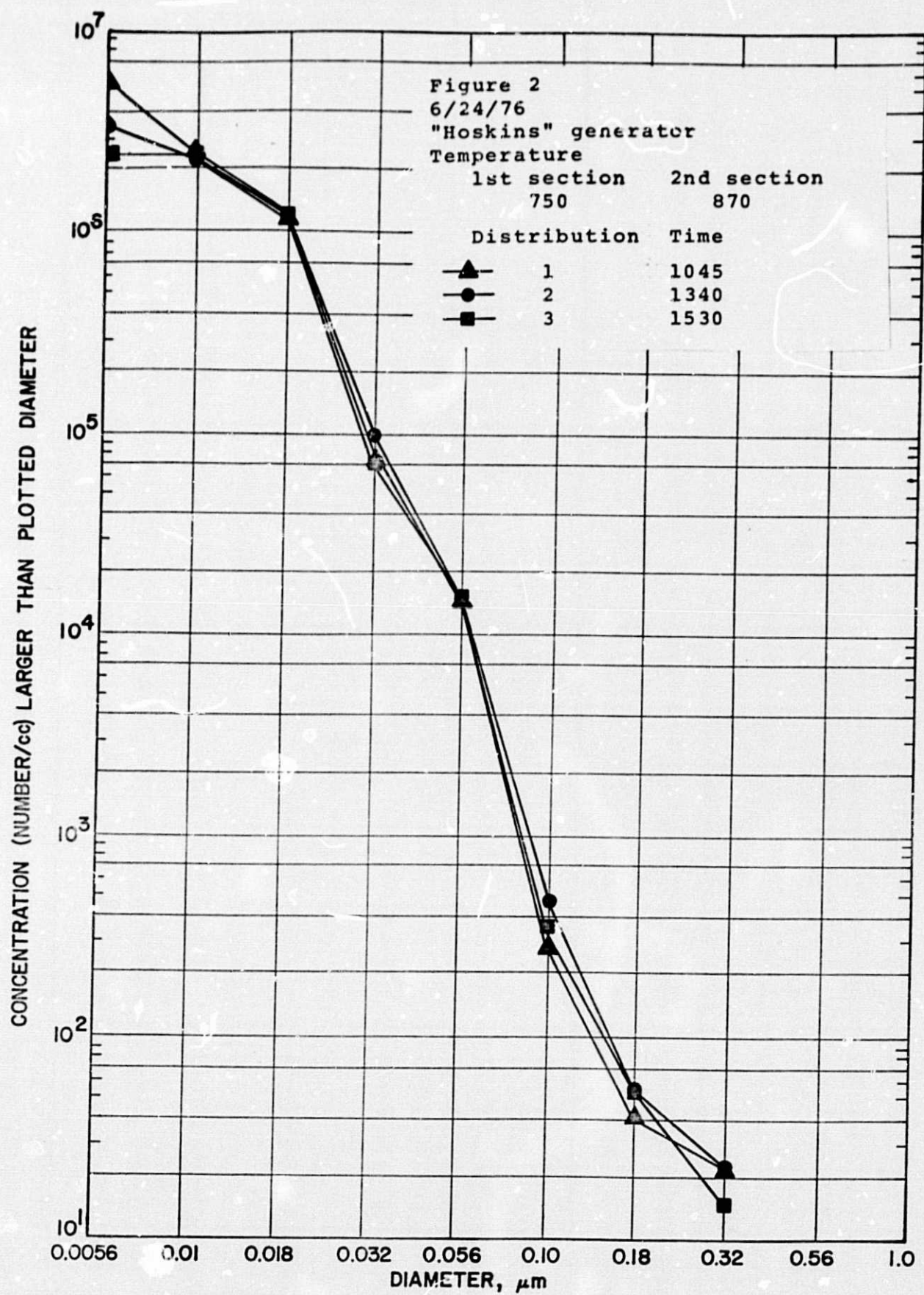


Figure 2

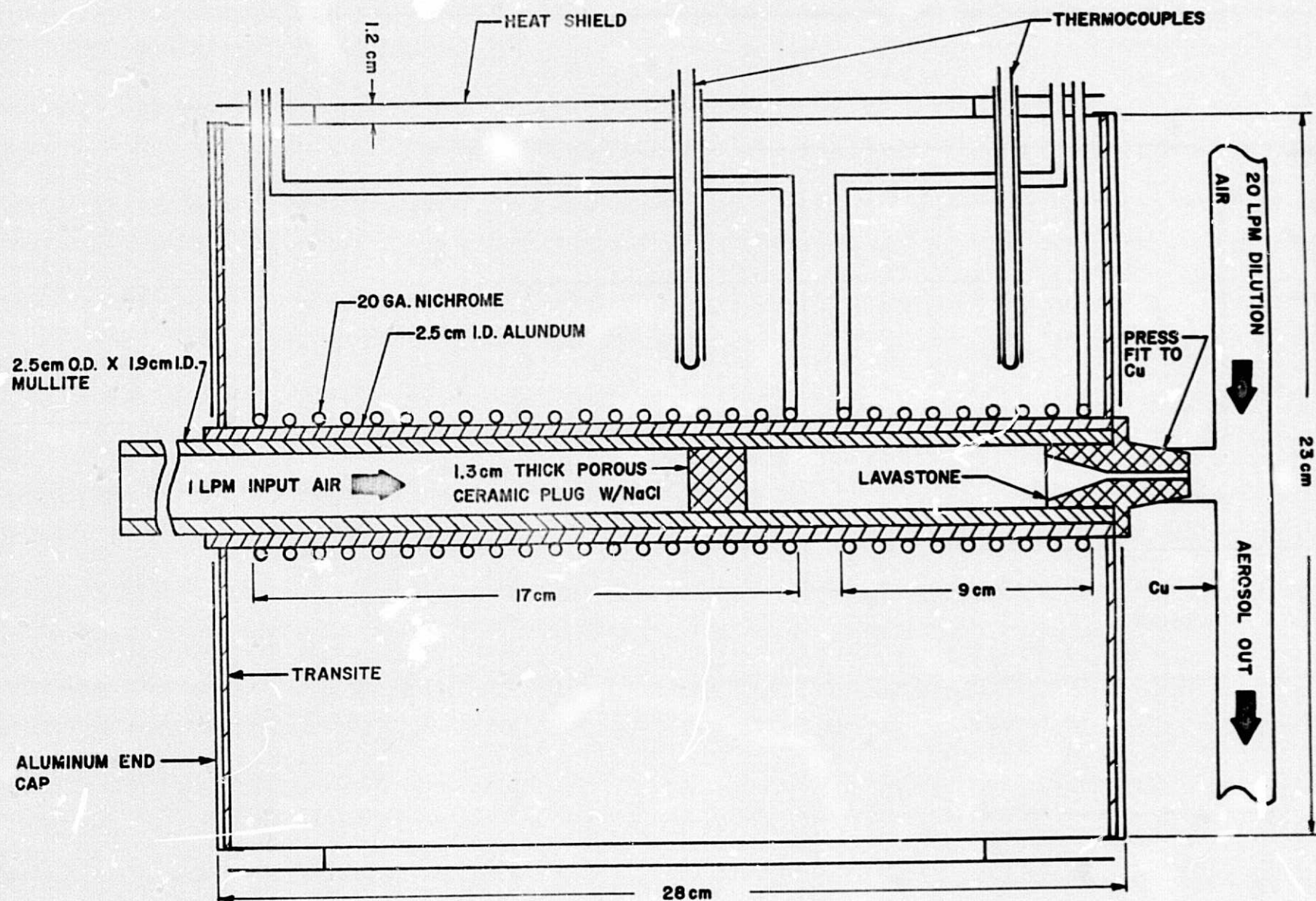


Figure 3

apparatus together.

The salt sources were contained in 2.5 cm O.D. x 1.9 cm I.D. mullite tubes which were inserted in the furnaces. These sources were 1.9 cm diam. by 1.3 cm long plugs cut from porous firebrick which was reportedly 98% Al_2O_3 , SiO_2 with traces of Fe_2O_3 , TiO_2 , Na_2O and K_2O . They were pressed into the mullite and cemented in place with Sauerbrey #1 paste. Between 2 and 3 ml. of a saturated NaCl solution were slowly added to these plugs which were then allowed to dry.

A lavastone plug, with a 1 cm O.D. x 0.5 cm I.D. tapered neck, was machined to fit into the exhaust end of the mullite tube. After firing, this plug was cemented into the tube and the neck pressed or cemented into a 1 cm I.D. copper tee. Again, one arm of this tee went to the dilution air while the other arm was attached to a coil of 3 m of 0.64 cm copper tubing. The purpose of this coil was to allow the heated generator air to cool before it entered any other part of the system.

The first prototype was constructed, as outlined above, in the shell from a discarded 36 cup coffeepot. The heater core, 25 cm long, accommodated two separately controlled heater sections: the source heater, 16 cm long, and a second heater 8 cm long. Unlike the first generator, this one had no condenser section between the two heated sections.

During part of the time this generator was in operation we had on loan from the manufacturer a Thermo-Systems model 3030, electrical aerosol analyzer (EAA). At that time we made a

comparison of the size distributions determined with the EAA and samples collected for electron microscopy. The results of these comparison tests are shown in figures 4-6. While the results of these tests are discussed later, the figures show that highly dispersed aerosols were produced with this generator.

The second prototype again consisted of two physically separable heated sections. These sections could either be separated to provide a condenser section between the two or bottled together so there was no cool region. The first section was 21 cm long while the second section was 11 cm long. Much of the data discussed later were obtained with this generator.

The final prototype, figure 3, returned to the concept of one unit with two separately heated sections. The total power required was less than 300 watts at 115V, 60 Hz. The primary difference between this unit and the two other prototypes was the use of thermocouples with stainless steel sheaths which were inserted in wells in the side of the generator. In the other prototypes, unsheathed thermocouples were inserted from the ends of the generator.

B. Temperature Measurement and Control

All temperature measurements were made with Chromel-alumel thermocouples. As shown in figure 1, one thermocouple for each heated section was used for both measurement and control functions. In the original generator, the thermocouples were placed between the mullite tube and the inside of the heater

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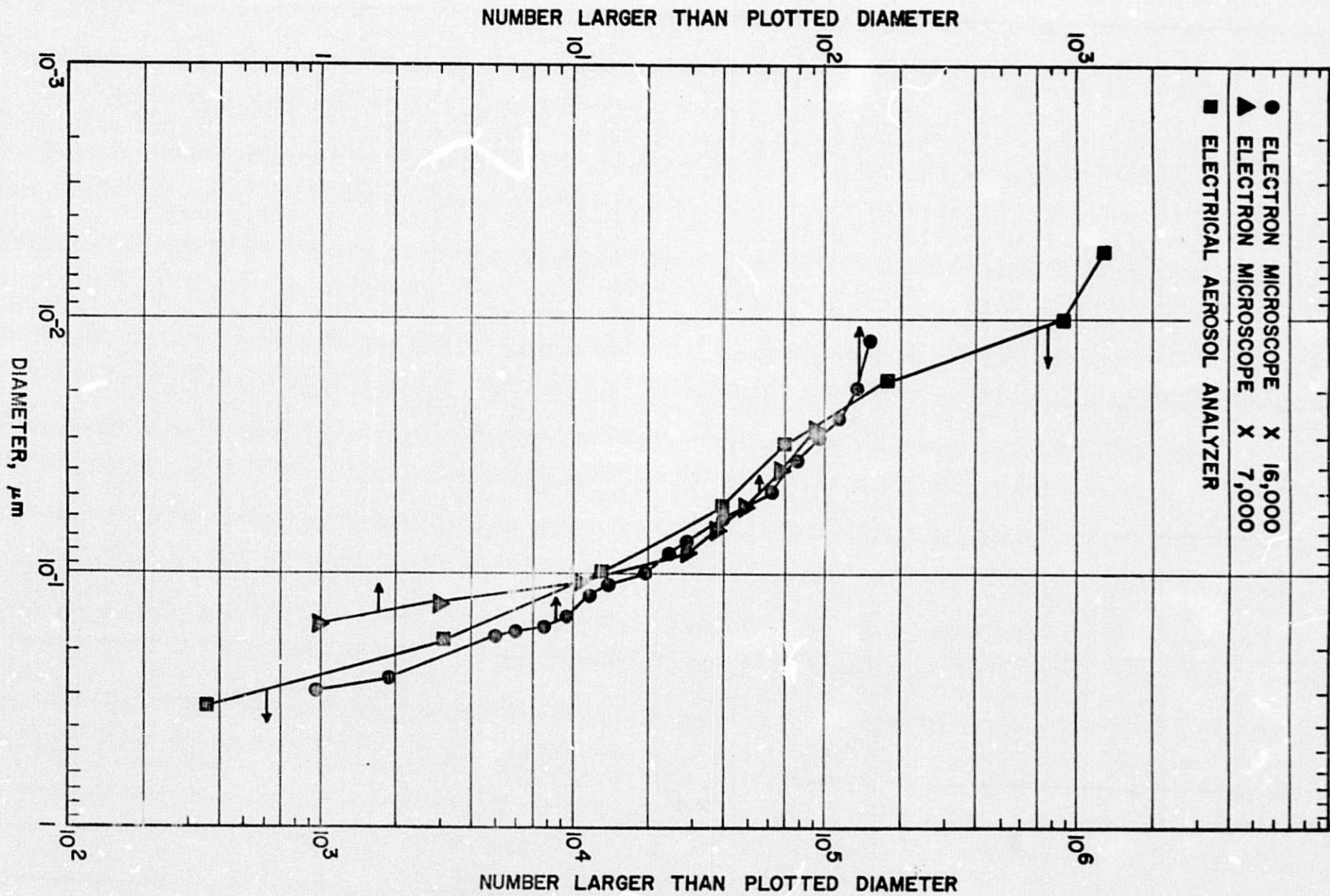
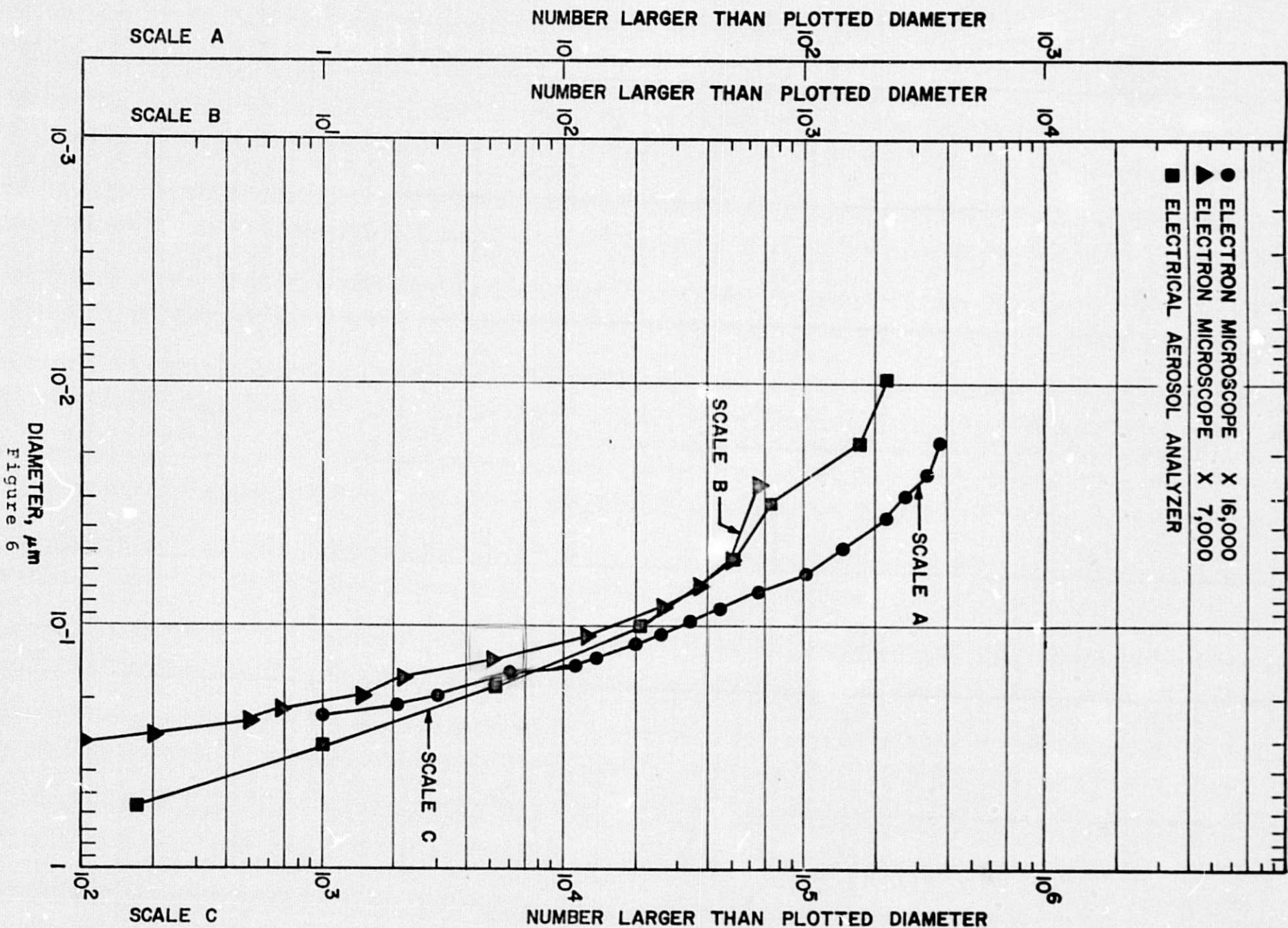


Figure 4





core. Unfortunately at high temperatures the electrical resistance of the heater cores decreased and the thermocouple outputs were quite noisy. While the outputs could be read to 0.1 mv the noise in these signals made the controllers unuseable. The power to these heaters was adjusted by setting the voltage applied with variacs.

In the three prototype furnaces, the thermocouples were placed outside the heaters. In these cases, even more than with the original generator, the measured temperatures were far different from the temperature within the mullite tubes. As an example, a number of actual temperature profiles were measured in the second prototype by pulling a thermocouple slowly through the mullite tube. During the time these profiles were being measured input air was passing through the mullite tube but the porous plugs were not in place. In the table below, the maximum temperatures which were measured in the two sections are compared with the readings of the fixed thermocouples for two of these profiles

<u>Generator Configuration</u>	<u>Section Max.</u>	<u>#1 Fixed</u>	<u>Section Max.</u>	<u>#2 Fixed</u>
Two sections separated by 7.8 cm	770	725	905	825
Two sections bolted together	785	700	885	760

As can be seen, the actual temperatures are much higher than the temperatures measured with the fixed thermocouples. While

these differences might be decreased by different placements of the fixed thermocouples, we believe a large discrepancy would still exist. Further, the introduction of the porous plug should have a large effect on the air temperature. As an example, figure 7 shows two distributions which are quite similar. Absolute concentrations should not be compared but only the shape of the size distribution. Curve A was obtained with the two heated sections of the second prototype bolted together. Curve B was obtained with the two sections separated by approximately 8 cm. As can be seen, the control temperature of the first section which contains the source, was over 200° hotter when the sections were separated than when they were bolted together. Although, as noted later, the source temperature would probably be higher under these conditions, it is unlikely it would need to be 200° higher.

In view of the above, the fixed thermocouples have been used to set and control the furnace temperatures to values which will produce the desired aerosol. These values once determined for a given generator configuration do not vary markedly from charge to charge. With further development it might be possible to seal thermocouples into the center of the mullite tube but these would need to be resistant to attack by hot NaCl and preferably of low thermal conductivity. At the present time it appears this is a needless refinement.

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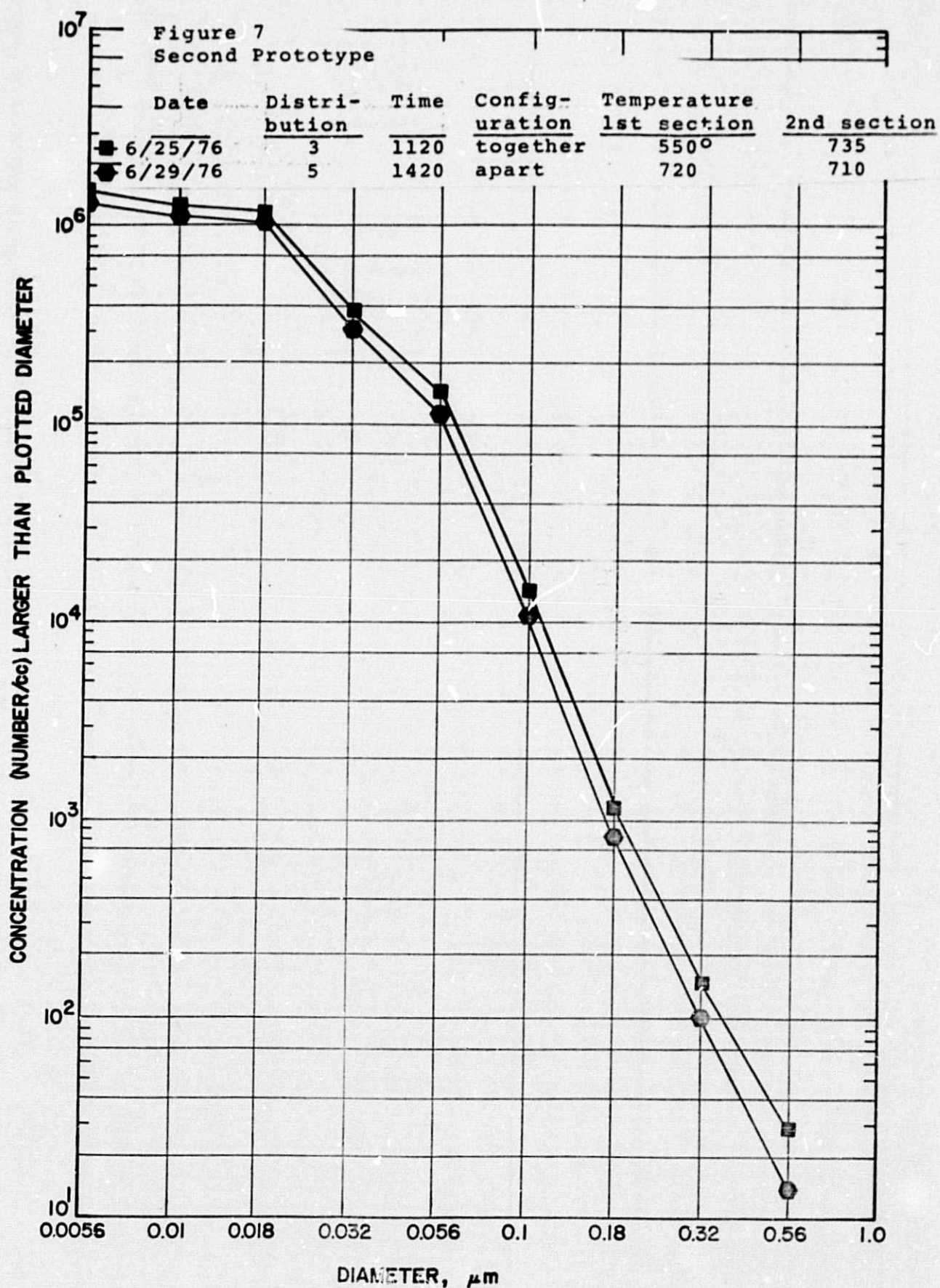


Figure 7

C. Aerosol Analyzer

During the first part of the contract period, size distribution and concentration data were obtained by collecting aerosol samples with an electrostatic precipitator for examination with a JEOL 7A/120 transmission electron microscope. Three major difficulties were apparent with this method. First, it was exceedingly time consuming. Second, there was a possibility that the samples being collected were not representative of the aerosol being generated. Finally, the opacity of the NaCl aerosol particles to the electron beam is insufficient to allow accurate size measurements of particles smaller than a few hundredths of a micrometer.

During December and January we had on loan, for about one week, a Thermo-Systems, model 3030, electrical aerosol analyzer (EAA). At that time we made a comparison of size distributions obtained with this analyzer and the electron microscope. These data are shown in figures 4-6. As can be seen, while the distributions determined by the two methods are not identical, they are quite similar. (Again, absolute concentrations cannot be compared.) When the technician who was working on this contract, Mr. George M. Hale, left in January to take a teaching position at West Texas State University his remaining salary and overhead money was converted to the purchase of one of these analyzers. With the exception of the electron microscope data in figures 4-6, all aerosol data presented in this report were obtained with this instrument.

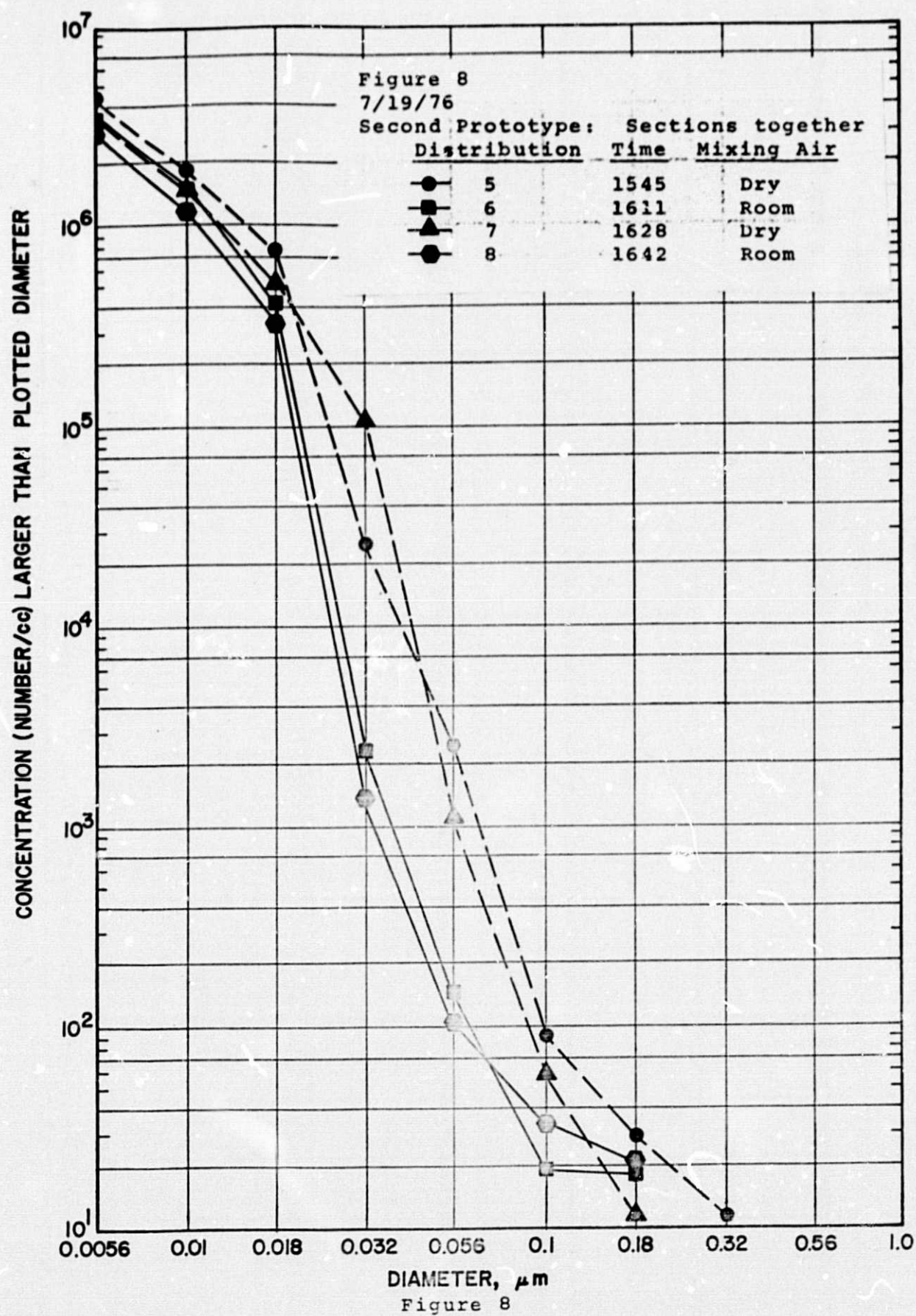
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While the instrument was received on April 3, it had to be returned to the factory for repair of a manufacturing discrepancy. At that time a modification was incorporated which supposedly makes the data for particles $0.01\text{ }\mu\text{m}$ and smaller more reliable. It was finally put back in operation the first part of June.

The total volume air flow required for the EAA is 50 lpm. Of this total 4 lpm is required for the aerosol analysis and 46 lpm for a sheath air flow. Further the temperatures of the aerosol and sheath flows should not vary by more than a few degrees centigrade. To ensure that these two flows are at the same temperature, they should be taken from the same supply. As the total flow from the generator system was only 21 lpm, an additional approximately 35 lpm was mixed with the generator air in a mixing chamber. The mixing air which was originally used was from the same dry air source as that for the input and dilution air.

During the latter part of this contract it was discovered that use of this very dry air, for the sheath flow, caused the ionizer-charger section of the EAA to operate outside of the manufacturers specifications. (A discussion with the manufacturer indicated they were unaware of this problem.) Figure 8 shows the possible magnitude of this effect. The dashed curves are data taken when dry mixing air was used, relative humidity about 3%. The solid curves are for data using filtered room air which produced a relative humidity greater than 20% in the sheath air. As is evident from this figure, the very dry air produced an apparent size distribution with a slightly greater

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fraction of large particles. Also, the reproducibility appeared to be less than with the more humid air. While there is always the possibility that humid air will cause the NaCl particles to grow, these data suggest this did not happen. Data obtained on another project in this laboratory also suggest negligible, if any growth, at these humidities.

D. Diluter

The final subsystem which needs to be discussed is the diluter. Immediately on exiting the generator, the aerosol laden input air is diluted approximately 20 fold with dry dilution air. After this primary dilution step the aerosol concentration is usually still too large to be handled by the EAA. To reduce these concentrations a dilution system, consisting of an HEPA absolute filter and valve in parallel, was added. As the air enters this diluter, part passes through the filter. Immediately downstream these two gas flows are recombined. The valve allows the fraction of the incoming air which passes through the filter and thus the final aerosol concentration to be controlled.

While this diluter appeared to work very well, at the very end of the contract period it was found that it could have a marked effect on the size distribution. Figure 9 shows two distributions using different dilution ratios. In an attempt to understand why this should be, other dilution schemes were tried. It was found that the measured size distribution changed with the method and amount of dilution.

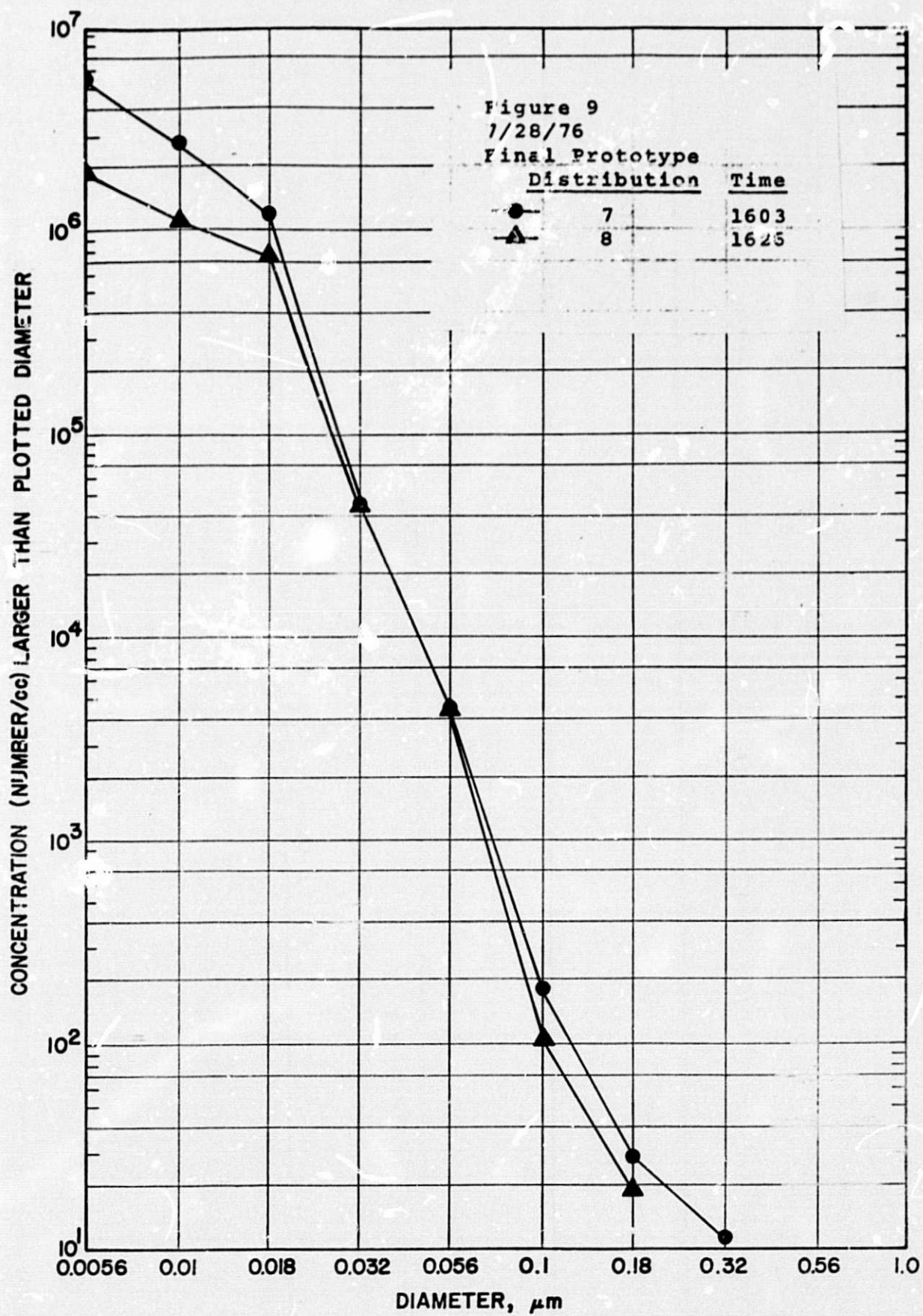


Figure 9

We have no reasonable explanation for this dilution effect. Neither coagulation and/or impaction appear capable of explaining it. The possibility that the aerosol was being charged was investigated by placing an aerosol deionizer at various places in the system. There were no appreciable differences in the distributions with and without the deionizer. While we cannot explain this dilution effect we have no reason to believe the measured distributions are in error. Thus, while this phenomenon needs to be further investigated, we believe the data we have obtained are reliable, within the accuracy limits of the EAA, for the generator-dilution system.

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IV. DISCUSSION

Rather than reproduce all of the quite large number of size distributions which have been obtained, we will only present examples which illustrate the operation of the generator system.

As can be seen in the data already presented, aerosols of the proper size distribution can be produced. Unfortunately it is still impossible to deliver, on demand, any one exact distribution or to predict, with confidence, what the distribution will be. Unless noted otherwise, the following discussion applies to all three prototype generators.

When a new generator is first used the approximate operating conditions are determined with the first salt charge. By charge is meant a mullite tube with lavastone exhaust and porous ceramic plug which has been treated with NaCl solution. It has been found that once having been used the ceramic plug is no longer porous. To make a new charge, the lavastone exhaust is sawed off and the ceramic plug removed, after which a new plug and the lavastone are cemented back in place.

With the second charge in place and the temperature controllers and input air flow volumes set to the appropriate values, aerosol first appears approximately one-half hour after supplying power to the furnaces. This aerosol consists of very small particles, generally smaller than $0.02 \mu\text{m}$. As the furnaces continue to heat, the particle concentration increases very rapidly. Because of this increase, the amount the aerosol stream is diluted must be increased so that changes in the

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measured distributions reflect the combined change of the generator and diluter. By the time the thermocouple outputs correspond to the control temperatures, approximately 45 to 60 minutes, the measured distributions are much flatter. That is, the larger particles constitute a greater fraction of the total aerosol population. In fact, these aerosols usually do not meet the required size distribution specifications. With time, although the total concentration may continue to increase slightly, the distribution becomes steeper so that within another hour, at most, the distributions meet specifications. Also by the end of this time, while the aerosol may be changing slowly, the rate of change is much less.

A possible explanation of this time history is as follows. When the salt first starts to get hot its vapor pressure is very low, the total amount of material available for aerosol formation is small, and consequently very small particles are produced. As the temperature increases, the amount of vapor increases and the particles grow larger. As the mullite tube and porous plug approach constant temperature the NaCl vapor density should approach a constant value. However, there is a temperature gradient across the inlet air and porous plug, the hottest portions being next to the tube walls. Any salt which is next to or on these walls will exhibit a higher vapor pressure than that further from the wall and will be depleted faster. In charging the porous plug, some NaCl solution usually managed to run down the walls of the mullite tube toward the inlet end. This salt, as well as that on the periphery of the plug, will

contribute to higher vapor densities when the furnace first reaches operating temperature than later when much of this salt has been depleted.

After these rather rapid initial changes, during which time the aerosol is probably useless for the purposes envisioned for it, the rate of change decreases markedly. The data for figure 10 were obtained with the second prototype with the two heated sections 9 cm apart. With the two furnace sections bolted together, so there was no intermediate cool section, the data of figure 11 were obtained. Data from the third prototype are shown in figure 12.

From figures 10 and 11 it is seen that to obtain the same approximate aerosol distribution, the measured temperature of the first section, that with the salt, needed to be much higher when there was a cool section separating the two heaters than when there was not. This is to be expected as much of the salt vapor will condense out in this cool section and not be available for aerosol formation at the exhaust. While we do not have data to prove it, we believe that with a cool center section the generator output may become more stable at an earlier time than if this section is absent. However, this attribute, if real, may well be balanced by the greater size and power consumption required for this configuration as well as by the more rapid depletion of the available salt.

Three difficulties were encountered whenever an attempt was made to determine the effect of either operating temperatures or input air flow volume on the aerosol which is produced. First, some time had to elapse after any change was made before

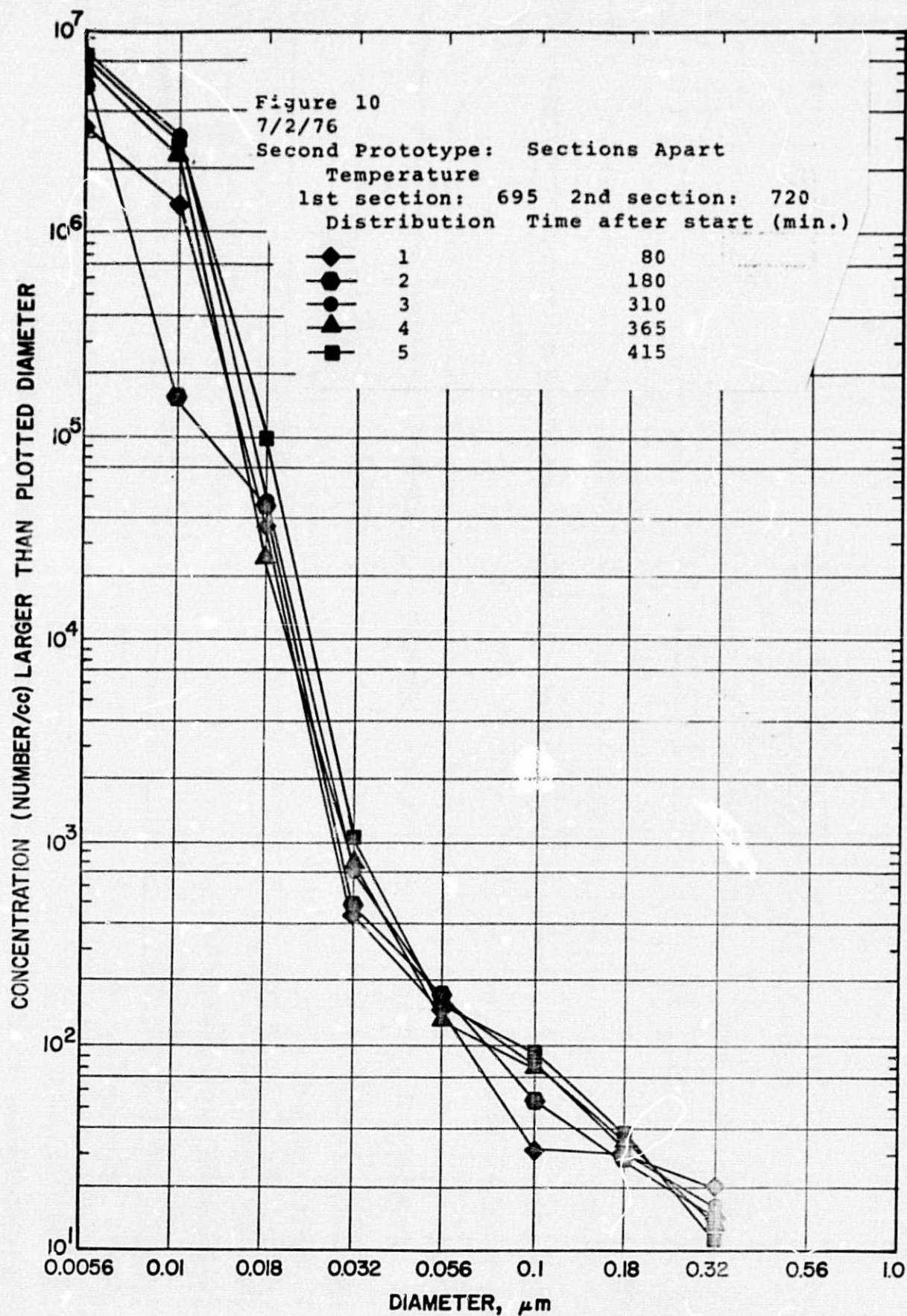


Figure 10

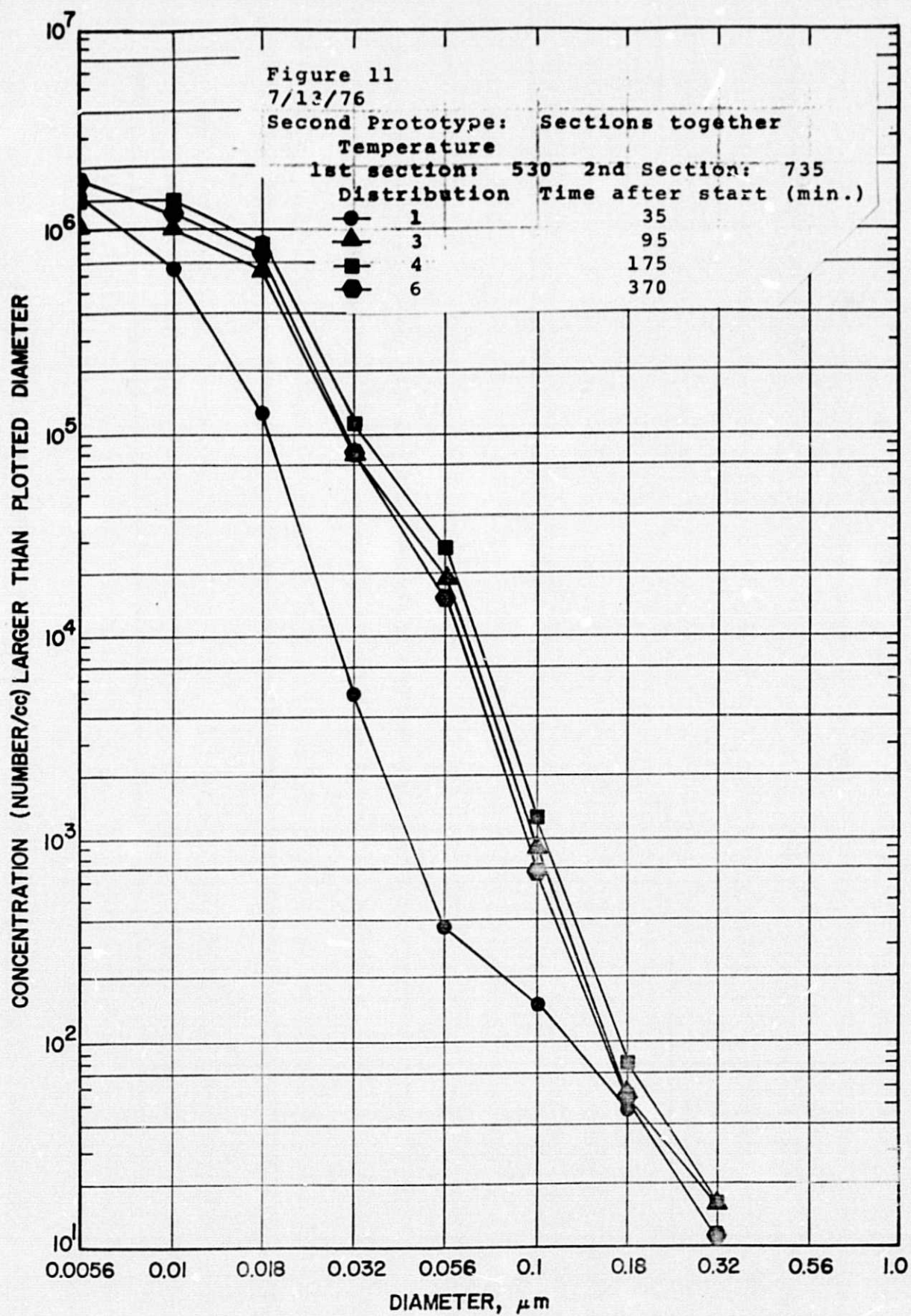


Figure 11

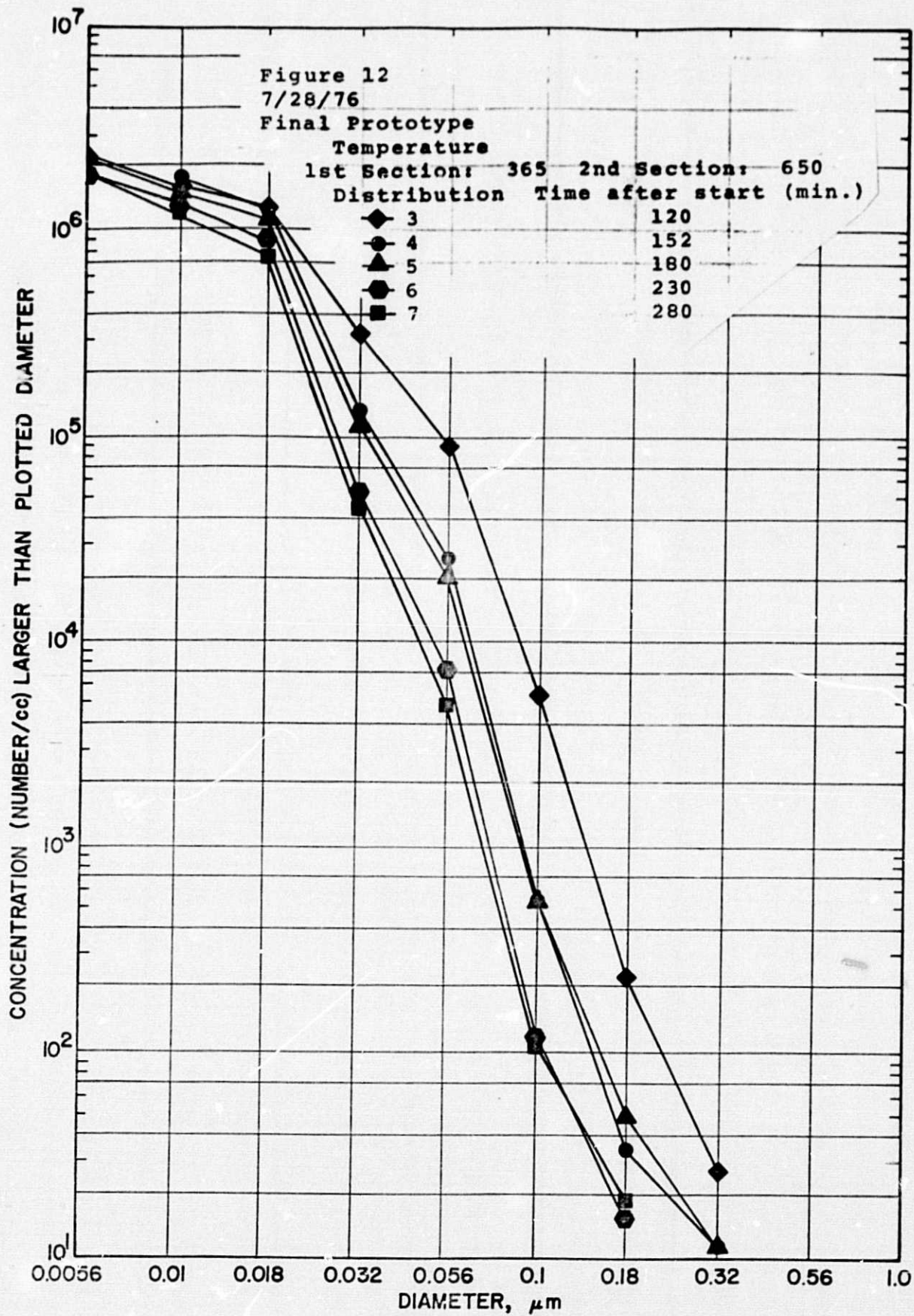


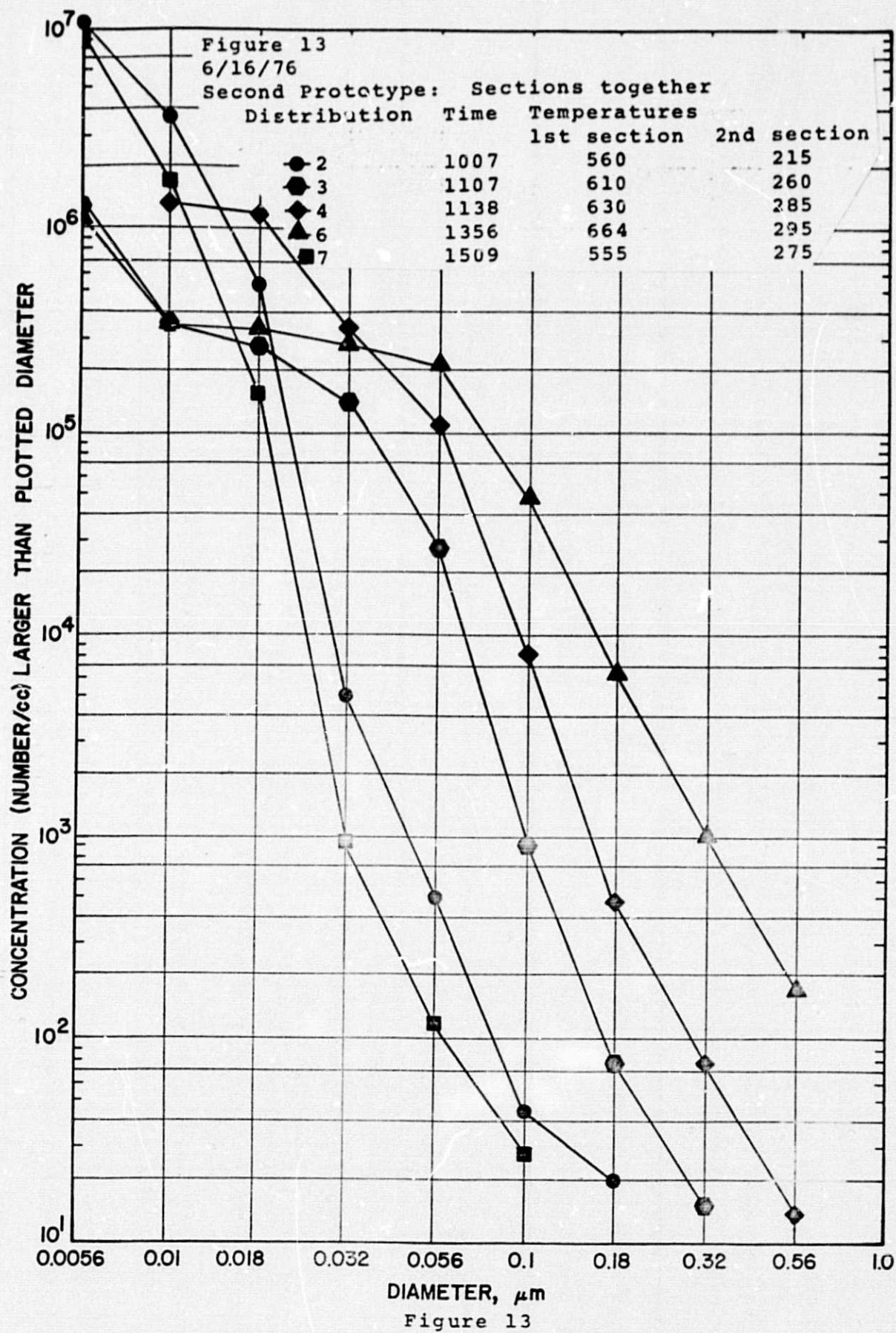
Figure 12

new equilibrium conditions were established. Unless the aerosol was absolutely stable before this change was made, it was sometimes difficult to decide how much of the change in the distribution was due to the altered operating conditions. Second, a change in any one temperature might well have had an effect on the other, and a change in air flow almost certainly affected the temperatures inside the mullite tube, even though the measuring thermocouples showed no change. Finally, if the total aerosol concentration changed markedly, the amount of dilution which was required changed which probably altered the distribution.

With the above in mind, we can make the following generalizations. Increasing the temperature of the first section flattens the size distribution. In figure 13 are shown data taken with the second prototype with the sections bolted together. In an attempt to minimize the effect of the second section, this heater was not turned on while these data were taken. The measured temperature of this section varied from 215° to 295° , well below that at which NaCl has an appreciable vapor pressure. Although the inside of this section must have been considerably hotter than the thermocouple indicated, the similarity of the first and last distributions would imply little effect from this section. As can be seen, higher source temperatures produced more larger particles as would be expected.

Unfortunately, we have no good data on the effect of the temperature of the second section. However, we have noticed no effects which could not be explained by other factors.

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Increasing input air flow volume also appeared to flatten the size distribution as shown in figure 14. These data were taken with the first prototype generator. Since increasing air flow should, if anything, cool the salt charge, a change in the temperature of the salt cannot explain these changes. Although different air flows could cause different temperatures in the second section, it is unlikely that they would differ enough to cause the sizeable changes in the distribution which were seen. It appears most likely that either there is excellent transfer of NaCl from the porous plug to the inlet air, at least at 1 lpm, or there is a marked change in the aerosol formation process as the gas cools on exiting the second section. One final possibility is a change due to different dilution ratios as the dilution was the same for the 2 and 4 lpm data while it was decreased at the time the 1 lpm data were taken. However, if dilution is the cause this is by far the largest change we have seen which can be attributed to this factor. Whatever the reason, we believe that control of the input air is the best method of changing the size distribution. This also means that control of the input air flow is important in the operation of the generators.

As to possible effects of dilution air volume, we again have no data to present. Because every other method of dilution appears to affect the distribution, we might expect the dilution air volume would also. Unfortunately we have not investigated this parameter. A volume flow rate of 20 lpm has been used throughout this work.

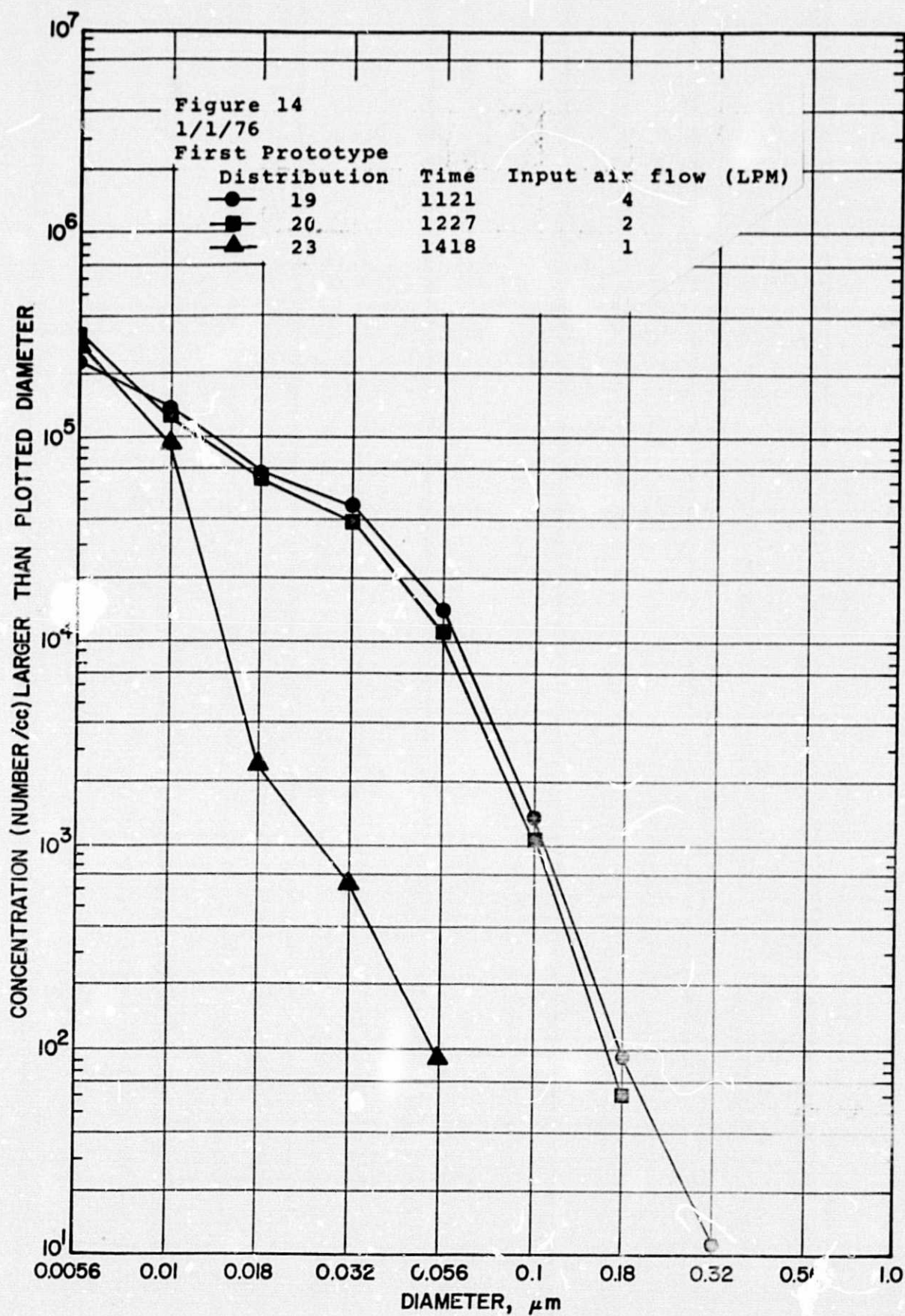


Figure 14

As noted in the introduction, condensation nuclei, CN, are often purposely introduced into the inlet air to provide sites for the condensation of NaCl vapor. The effect of the presence of CN was investigated early in the contract using the very first generator and the nuclei produced by a heated chromel wire. The aerosols were examined with the electron microscope. There were no apparent differences in the aerosols generated with and without CN.

With the second prototype, a few runs were conducted in which the mullite tube, lavastone exhaust, and porous plug were pretreated by heating to at least 1000°C until fewer than 100 CN/cc were emitted at normal operating temperatures. The plugs were then charged with salt and aerosols generated. Unfortunately the temperature controllers were not operating properly during this time so it did with the non-pretreated tubes. This The only difference noticed was that with the pretreated tubes the lavastone exhaust seemed to become plugged with NaCl more readily than they did with the non-pretreated tubes. This could have indicated the presence of CN, with the non-pretreated tubes, which promoted condensation in the gas phase and reduced the amount of salt deposited on the walls.

Finally, we would like to present some data on the coagulation of the highly dispersed aerosols produced with these generator systems. These data were obtained by first filling a 0.7 cm^3 ($0.7 \times 0.7 \times 1.6\text{ cm}$) aluminized mylar bag with aerosol. This took approximately 10 minutes. Because the temperature of the aerosol had a greater time to equilibrate with the

temperature of the air in the room, the size distribution data were obtained by connecting the bag only to the aerosol inlet, the sheath air being taken from the room. Consequently, only a small fraction of the total aerosol was used and the bag volume and geometry remained fairly constant. Wall losses in this system are assumed negligible compared to the losses from coagulation.

Figures 15 and 16 present the data from two experiments with aerosols produced with the final prototype generator. As was expected, coagulation proceeded rapidly at first, especially with the smallest particles. With time the concentration of these particles decreased until after approximately two hours there were few, if any, particles smaller than $0.02 \mu\text{m}$. At the same time the concentration of larger particles was increasing but at a much slower rate than the small particle concentration was decreasing. Again, these changes were to be expected. At the end of 18 hours there were few particles smaller than $0.056 \mu\text{m}$ while the concentration of particles larger than $0.56 \mu\text{m}$ had increased by a factor of about three. The maximum concentrations of the larger particles occurred some hours after the bag was filled but before 18 hours. This could well be explained by continued coagulation involving the larger particles and the settling of these particles combined with a decreased rate of formation due to smaller concentrations of the smaller sizes. However, it should be noted that the largest particles measured by the EAA are $1 \mu\text{m}$ and above $0.32 \mu\text{m}$ the reliability of the measurements decreases. The analyzer actually measures the charge carried by the aerosol and the larger the

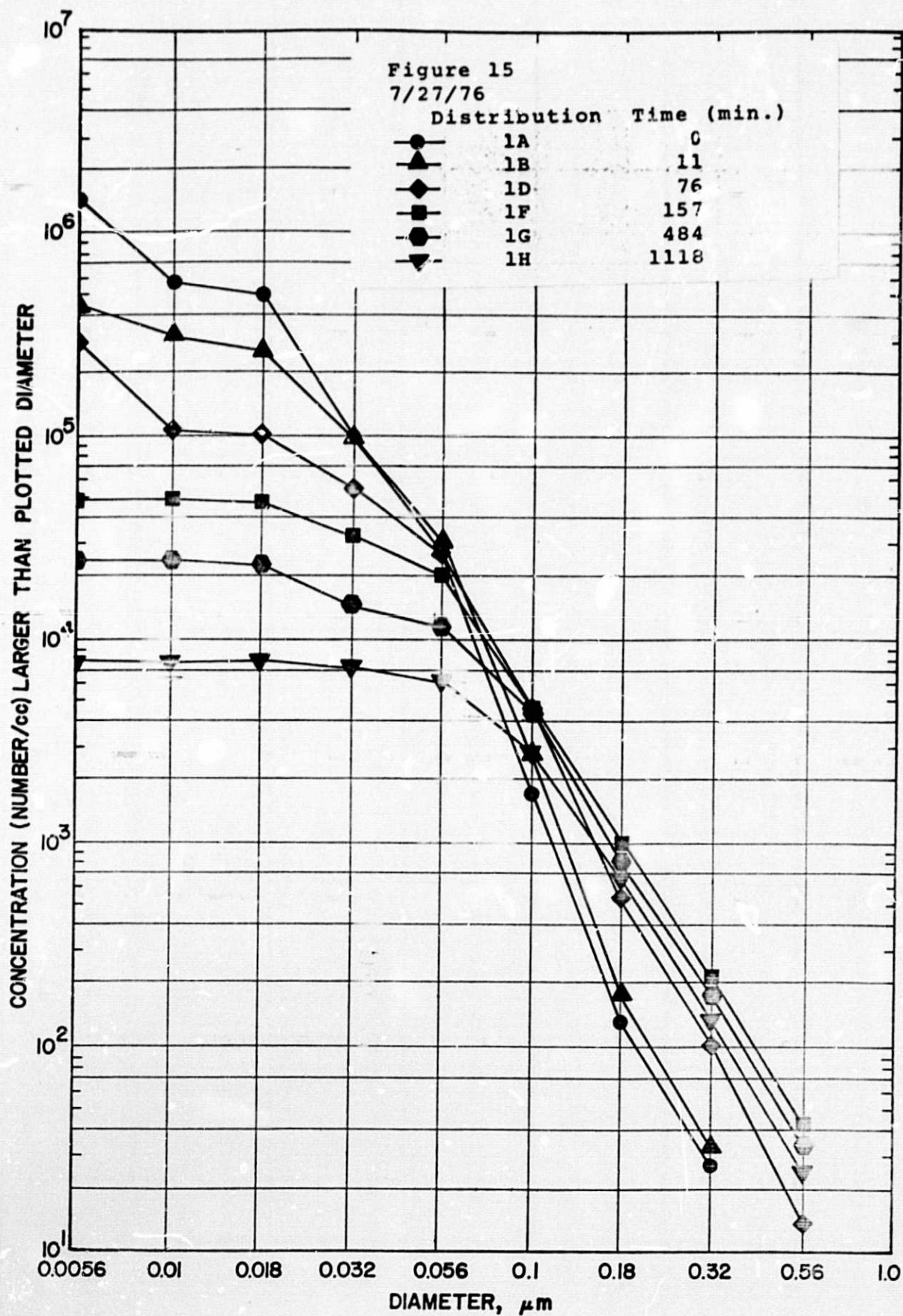
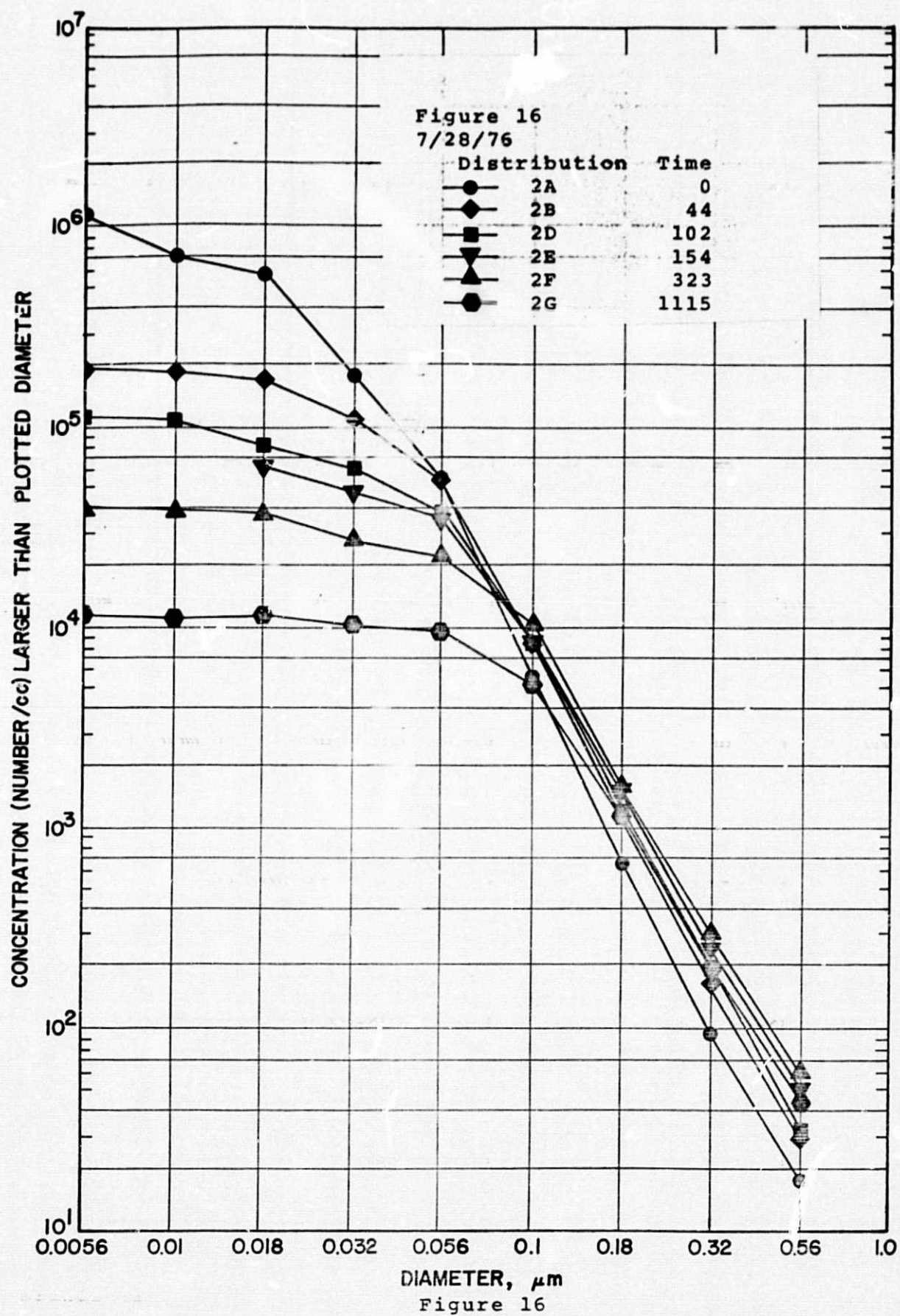


Figure 15



particle the greater the charge which may be carried. Thus, a greater degree of caution is required when interpreting data at the large end of the distribution than in the range between 0.01 and 0.32 μm .

The similarity of these two sets of data and the fact that the gross changes are what would be expected give us an added measure of confidence in the reliability of the EAA data. Unfortunately, it also indicates that storage of these aerosols for any length of time will produce distributions unsuited to the needs of the experiments for which the aerosols are to be used. There are four possibilities for overcoming this storage problem. First, it might be possible to generate an aerosol with small enough particles so that even with higher coagulation rates there would be sufficient time to fill a container and then remove the aerosol for the desired experiments. While more data are needed to determine whether this is possible, at the moment it appears this is not a valid solution because of the exceedingly high coagulation rates. Another possibility is to remove by impaction or filtration, such as with nucleopore filters, the large end of the distribution when the aerosol is removed from the storage container. The most obvious answer is to store the aerosol at much lower concentrations. Unfortunately, from the standpoint of our system, the concentrations could be so low that the EAA could not supply much size information. However, other methods, such as a CFD, might be used to obtain these data. Finally, if we can understand the dilution effect so as to be able to predict how the distribution will change with dilution or find a dilution

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method which does not alter the distribution, we could fill the storage container with a known aerosol at much lower concentrations than is now possible. Still, even if a known, diluted aerosol is stored, there is still a great need for data on the coagulation of highly dispersed aerosols. If this information is not available there is no assurance that an aerosol removed from the storage container will meet the requirements of an experiment even though the original size distribution and concentration were known.

V. SUMMARY

Based upon aerosol size analyses using a Thermo-Systems, Inc., model 3030, electrical aerosol analyzer, highly dispersed sodium chloride aerosols can readily be generated by vaporization-condensation techniques. We have built and tested four generator systems all of which produce the required size distribution. This distribution is one in which the concentration of particles between 0.02 and 0.1 μm diameter is 500 times the concentration of particles larger than 0.1 μm .

Three of these generators were constructed with the requirements of the Shuttle/Spacelab in mind. They all utilized electrically heated tube furnaces and replaceable mullite tubes which contained the salt charges. These charges were porous ceramic plugs, glued into the mullite tubes, which were treated with a NaCl solution and dried. Approximately 21 lpm of air were required for generator operation. The final prototype furnace was 25.4 cm O.D. by 30.5 cm long and required approximately 300 watts of electrical power to maintain the required temperatures.

All generators had two separately heated sections. In some cases the heated sections were separated by a cool section and in other cases the two sections were either contained in the same furnace or were bolted together. Temperatures were measured and controlled with chromel-alumel thermocouples which were usually placed outside the heating elements. Because of this placement, the actual temperatures within the mullite tubes were unknown. However, it appears that a salt temperature of

about 775° is sufficient to produce the desired aerosol.

The general characteristics of these generation systems are outlined below.

1. It takes 1-1/2 to 2 hours before the size distribution becomes moderately stable. Even then there may be slow changes with time.

2. One salt charge will produce aerosols for at least two consecutive eight hour periods.

3. Increasing the amount of air which flows through the generator increases the fraction of large particles. Control of this air flow appears to be the best method of controlling the size distribution.

4. Increasing the temperature of the salt source also increases the size, and number, of the particles.

5. The amount the aerosol is diluted between the time it leaves the generator and reaches the electrical aerosol analyzer appears to effect the size distribution and concentration.

The electrical aerosol analyzer which was used to obtain the majority of the size distributions appears to provide reasonably reliable data. While we cannot verify the absolute accuracy of these distributions they compare favorably with distributions obtained with samples collected with an electrostatic precipitator and analyzed with a transmission electron microscope.

Finally, when these aerosols were stored at total concentrations of about 10^6 /cc, coagulation of the smallest particles proceeded very rapidly.

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VI. CONCLUSIONS

The generators we have built produce more than enough aerosols of the specified size distribution and, we believe, are applicable to the Shuttle/Spacecraft environment. However if the size, weight or power or air requirements are excessive, the generator can probably be scaled down. If this is done, the stability and/or the length of time one salt charge will last may suffer.

While the size distribution is readily obtained, producing the proper concentrations may be a greater problem if the dilution effect we have noticed is real. Any vaporization-condensation generator will undoubtedly produce concentrations much larger than the 10^2 to 10^3 /cc which is required. This makes some form of dilution necessary. There also needs to be some way of controlling the concentration within the range 10^2 to 10^3 /cc. For these reasons the effect of the method and/or amount of dilution on the distribution needs to be known.

Although we do not know the absolute values, in our system the concentration of particles in the aerosol when it entered the diluter must have approached or exceeded 10^8 /cc. There is a possibility that at lower concentrations there would have been little, if any, discernible effect of the diluter. If this is so, the dilution effect is of less importance.

If it is decided to store the aerosol before use, the change in the distribution with time, due to coagulation, needs to be determined. The data presented earlier show rapid coagulation at rather high particle concentrations, 10^6 /cc. At lower

concentrations these rates would be much slower. However, until the actual coagulation behavior of these particular aerosols is known, there is a possibility the rates may still be sufficiently high, at the concentrations which would be stored, that the aerosol removed from the storage container will not be suitable. The EAA requires concentrations approaching 10^5 /cc to obtain reasonably reliable data. To study coagulation at much lower concentrations will require either a more sensitive EAA or a different analytical technique. However, until such an instrument is available for these studies, data obtained at the higher concentrations would be valuable for verifying present coagulation theories and/or formulating expressions which would describe and predict the actual coagulation of these highly dispersed NaCl aerosols.

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